Time-Resolved Investigation of Exciton-Polariton Dephasing in CuCl

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The intrinsic dephasing time of an exciton polariton is investigated in the time domain by use of a time-resolved coherent two-photon excitation and probe technique. This method allows discrimination of the elementary interaction processes of the exciton polariton in semiconductors. The results obtained in CuCl show the role of the different phonons in the exciton-polariton dephasing.

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Propagation and redistribution of electromagnetic energy near the band gap of polar semiconductors is expected to proceed through the exciton polariton. It is thus of fundamental interest to have information on the underlying dynamics of these quasiparticles and, in particular, on the processes that condition their coherence and lifetime. These aspects up to now have been investigated using indirect techniques lacking selectivity for the discrimination of the exciton-polariton relaxation channels.

Thus, in the time domain, the incoherent time-resolved techniques,[1-3] such as time-resolved luminescence or induced absorption, only provide global information on the energy content and loss in the crystal, subsequent to a few scattering events averaging out the elementary scattering mechanisms. Furthermore, extraction of this information is complicated because of the inherent polariton propagation and transmission at the crystal surfaces which may substantially alter the bulk polariton features.[4] The exciton-polariton dephasing has also been addressed in the time domain using a time-resolved four-wave-mixing technique. This technique proceeds via strong linear polariton excitation within the absorption layer, close to the crystal surface, and, hence, only gives access to the high-excitation regime where the polariton dephasing is dominated by polariton-polariton interactions.[5,6] In the frequency domain, the dispersion of the polariton damping has been indirectly studied[7] but with a limited selectivity.

Here we show that the intrinsic dephasing of an upper-branch exciton polariton can be precisely and directly measured along its dispersion curve in a noncentrosymmetric semiconductor by a time-resolved two-photon coherent nonlinear technique. The demonstration of this new method, which allows the first direct determination of an exciton-polariton dephasing time, is performed on the Z_3 exciton polariton in cuprous chloride (CuCl) whose characteristics have been extensively investigated.[8] The technique consists of picosecond coherent two-photon excitation and detection of an exciton-polariton wave packet. Coherent excitation of an exciton polariton of frequency $\omega_{e\pi}$ and wave vector $\mathbf{k}_{e\pi}(\omega_{e\pi})$ is realized in the bulk of the crystal by two-photon absorption of two synchronized picosecond pulses with frequencies ω_1 and ω_2 and wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ such that $\omega_{e\pi}=\omega_1+\omega_2$ and $\mathbf{k}_{e\pi}=\mathbf{k}_1+\mathbf{k}_2$. These conditions can only be satisfied for the upper-branch polariton, restricting the applicability of the technique to this mode.[9] The exciton polariton is an admixture of a material excitation and an electromagnetic mode and so its coupling with the excitation fields occurs through both of these components. Close to the polariton stop band and with neglect of spatial dispersion, the amplitude of the coherently excited wave packet is proportional to

$$d_\pi = d_E(\omega_e^2-\omega_{e\pi}^2)+d_M\,, \tag{1}$$

where ω_e is the bare exciton frequency at $\mathbf{k}=0$ ($\omega_e \sim 3.202$ eV in CuCl). The coupling parameters d_M and d_E are related, respectively, to two-photon absorption and sum-frequency generation close to ω_e.[10] Interference of these contributions leads to a strong dispersion in d_π and its vanishing either above or below ω_e, depending on the relative sign of d_E and d_M. In the case of CuCl, this occurs around $\hbar\omega_{e\pi}=3.218$ eV,[10,11] very close to the polariton energy $\hbar\omega_{e\pi}^f=3.217$ eV, for excitation beams propagating along the same direction, i.e., $\theta=0°$ where θ is the angle between $\mathbf{k}_1$ and $\mathbf{k}_2$.

In the most general situation, when the local excitation process has terminated, the polariton wave packet will propagate inside the crystal with its group velocity $V_g(\omega_\pi)$ along a direction fixed by $\mathbf{k}_{e\pi}$. However, in the case of CuCl, relaxation occurs much faster than propagation and the wave packet can be probed locally. The evolution of the exciton-polariton coherence is followed by phase-matched parametric emission at $\omega_d=\omega_{e\pi}-\omega_p$ stimulated by a third picosecond pulse ω_p, delayed with respect to the excitation. For $\theta=0°$, the intensity of the coherent signal as a function of the probe time delay t_D can be readily obtained:

$$S_0(\omega_{e\pi},t_D)\sim|d_\pi|^4 e^{-2t_D/T_2}\int_{-\infty}^{+\infty}dt' e^{-2t'/T_2}|E_p(t')|^2\left|\int_{-\infty}^{t'}F(\tau)d\tau\right|^2, \tag{2}$$

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where $F(\tau)$ is a two-photon excitation function. Similar expressions can be calculated for $\theta \neq 0°$ with, in particular, the same t_D dependence. For long time delay t_D, i.e., for $t_D \gg t_p$, where t_p is the pulse duration, the amplitude of the excitation is constant and, hence, the signal decreases with a characteristic time $T_2/2$ as t_D increases, directly measuring the exciton-polariton dephasing time T_2. We emphasize that besides the temporal separation the excitation and probing stages could also be separated in space using the nonlocal possibilities of the nonlinear probing techniques.[12] For the long-living exciton polariton this should allow a direct investigation of both spatial and temporal evolution of the wave packet inside a crystal as demonstrated for the phonon polariton.[12]

The demonstration of this technique was performed on two upper-branch polaritons in CuCl with energies $\hbar\omega^b_{e\pi} \approx 3.208$ eV and $\hbar\omega^f_{e\pi} \approx 3.217$ eV corresponding, respectively, to a backward, $\theta = 180°$, and a forward, $\theta = 0°$, excitation geometry. The experiments were conducted using a passively mode-locked Nd^{3+}:glass laser delivering a single 5-ps pulse in the infrared ($\omega_I = 1.054$ μm) which is frequency converted to create the three independent pulses ω_1, ω_2, and ω_p. The pulse at frequency ω_1 is a small part of the initial infrared pulse while ω_2 is tunable around 611 nm; the probe-beam frequency ω_p, in the infrared, is adapted to phase-matching requirements and is either at the same frequency as ω_1 for $\theta = 180°$ or frequency shifted to 1.107 μm by stimulated Raman scattering in carbon tetrachloride for $\theta = 0°$. After polarization adjustment, the three beams are focused into a cooled 2-mm-long CuCl sample with (110) surfaces. This orientation was chosen to obviate the influence of the longitudinal mode and corresponds to a (110) polarization of the polariton.[11] The signal is detected by a photomultiplier after spatial, spectral, and polarization selection. In the 180° geometry, in order to discriminate the signal against the frequency-degenerate diffused beam at ω_2 a temporal selection of the signal is added using a Kerr gate synchronized with the probe pulse.

The results of the experiments for the two investigated polaritons $\omega^f_{e\pi}$ and $\omega^b_{e\pi}$, at a crystal temperature of 7 K, are plotted in Fig. 1 and evidence the strong dispersion of the dephasing time. As anticipated, the smallness of the signal at $\theta = 0°$ is a consequence of destructive interference between the material and electromagnetic contributions in d_π. Here the small-delay ($t_D \leq 10$ ps) signal due to the nonresonant third-order nonlinear response of the crystal is about 10^5 times stronger than the resonant one. This has precluded the observation of the $\theta = 0°$ polariton mode in previous two-photon absorption spectroscopy in the frequency domain.[9-11] In our case these two contributions are separated by their different temporal behaviors and an exponential decay of the polariton coherence is observed over nearly 4 orders of magnitude for $t_D \geq 10$ ps, yielding a dephasing time T_2 of 21 ± 3 ps at $\hbar\omega^f_{e\pi} \approx 3.217$ eV and $T = 7$ K. A

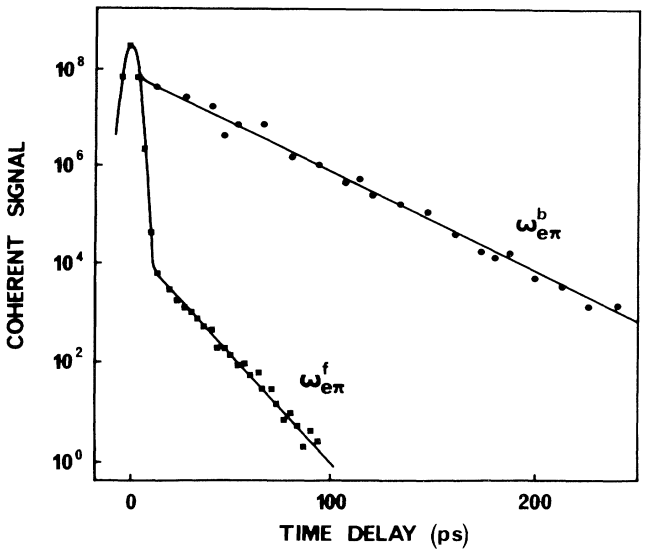

FIG. 1. Coherent signal measured in time-resolved two-photon excitation and probe experiment on $\omega^b_{e\pi}$ (upper curve) and $\omega^f_{e\pi}$ (lower curve) in CuCl at 7 K, plotted on a logarithmic scale as a function of probe delay (t_D).

similar behavior is observed for the $\omega^b_{e\pi}$ polariton but with a stronger resonant signal; the intensity ratio of the resonant signals, $I^f_R/I^b_R \approx 10^{-4}$, is in good agreement with the estimation of Ref. 10. A dephasing time T_2 of 43 ± 4 ps is measured for $\hbar\omega^b_{e\pi} \approx 3.208$ eV with a dynamics limited to 5 orders of magnitude by residual diffusion of the ω_2 beam.

At a low density of exciton polaritons, the dephasing is dominated by exciton-polariton scattering by spatial and temporal disorder of the lattice. In the case of CuCl, scattering by impurities, or other spatial disorder effects, is unlikely because of the smallness of the exciton Bohr radius and the weakness of the polariton group velocity.[6] The main dephasing processes are expected to proceed from phonons which temporally modulate the crystal order. The strong frequency dependence of T_2 is thus expected to reflect the variation of the exciton-phonon interaction along the polariton dispersion curve.[6] The relevance of this dephasing process can be directly evidenced in the present investigation.

The most probable processes for upper-branch polaritons are extraband down- and up-conversion into a lower-branch polariton with, respectively, emission or absorption of a phonon[13] (Fig. 2). These processes are strongly enhanced compared to intraband ones because of the higher density of final accessible states. At low temperature the relaxation is expected to be dominated by exciton-polariton scattering by acoustic phonons. In a zinc-blende-type crystal the interaction is mediated mainly by the deformation potential (DP) for the LA phonon and by the piezoelectric effect (PE) for TA phonons,[14] so that the low-temperature exciton-polariton

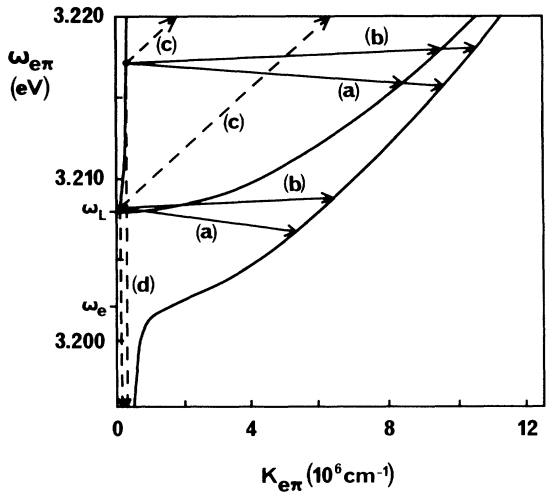

FIG. 2. Measured exciton-polariton and longitudinal exciton dispersion curves in CuCl (Ref. 16). The arrows indicate the phonon-assisted relaxation channels for the $\omega^f_{e\pi}$ and $\omega^b_{e\pi}$ upper polaritons. Processes a (c) and b (d) correspond, respectively, to polariton down- and up-conversion into a lower-branch polariton assisted by an acoustic (longitudinal) phonon. Polariton relaxation into a longitudinal exciton for the higher-energy polariton only leads to small modifications of the dephasing rates.

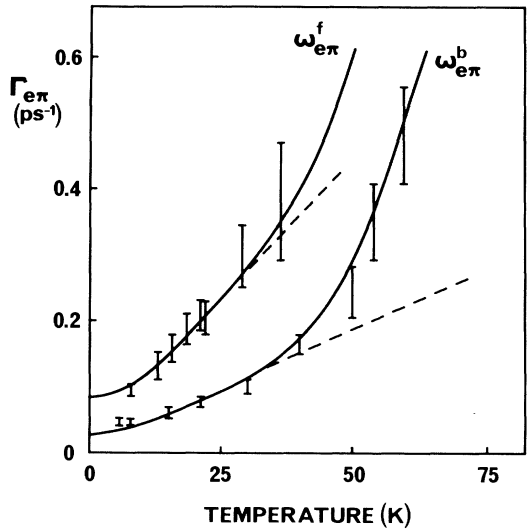

FIG. 3. Temperature dependence of the $\omega^f_{e\pi}$ and $\omega^b_{e\pi}$ polariton dephasing rate $\Gamma_{e\pi}=2/T_2$ (upper and lower curves, respectively). The dashed and solid lines are calculated using, respectively, Eqs. (5) and (7) (see text).

damping rate $\Gamma_{e\pi}=2/T_2$ can be written as

$$\Gamma_{e\pi}=\Gamma^{LA}_{DP}+\Gamma^{TA}_{PE}, \tag{3}$$

where Γ^{LA}_{DP} and Γ^{TA}_{PE} are the relaxation rates associated, respectively, with the DP and PE scattering mechanisms. The frequency dependences of these processes are[7,15]

$$\Gamma^{LA}_{DP}\sim q^2\sim\omega_{e\pi}-\omega_e, \tag{4a}$$

$$\Gamma^{TA}_{PE}\sim q^4\sim(\omega_{e\pi}-\omega_e)^2, \tag{4b}$$

where q is the wave vector of the final lower-branch polariton. This indicates that the role of the PE mechanism increases with $\omega_{e\pi}$. The theoretical damping rate is thus expected to increase with $\omega_{e\pi}$ in agreement with our experimental results. Discrimination between the two mechanisms can be realized because of their different temperature behaviors which are governed by the Bose factors of the involved phonons. These are different for the DP and PE processes and the temperature dependence of $\Gamma_{e\pi}$ is given approximately by

$$\Gamma_{e\pi}(T)=\gamma_{DP}[1+n(\omega^-_{LA})+n(\omega^+_{LA})]+\gamma_{PE}[1+n(\omega^-_{TA})+n(\omega^+_{TA})], \tag{5}$$

where γ_{DP} and γ_{PE} are frequency-dependent coupling parameters and $n(\omega)$ the occupation number for the ω phonon. The frequencies ω^+ and ω^- of the acoustical phonons involved, respectively, in the up- and down-conversion processes are imposed by energy and wave-vector conservation and can be determined using the acoustic-phonon data and exciton-polariton dispersion curves in CuCl.[16] The results of the dephasing-rate measurements as a function of crystal temperature are plotted in Fig. 3. The low-temperature (≤ 40 K) dependence of $\Gamma_{e\pi}$ can be reproduced with $\gamma^f_{DP}=37$ μeV and $\gamma^f_{PE}=25$ μeV for the $\omega^f_{e\pi}$ polariton and $\gamma^b_{DP}=14$ μeV and $\gamma^b_{PE}=3$ μeV for the $\omega^b_{e\pi}$ polariton (dashed lines in Fig. 3). The frequency dependence of the coupling parameters are in agreement with the theoretical estimations [Eqs. (4)] with, in particular, a faster increase of γ_{PE} with $\omega_{e\pi}$. We emphasize here that none of the temperature behavior of the damping rates can be reproduced with solely one mechanism. The efficiencies of the PE and DP mechanisms are thus comparable in CuCl, as observed in the related CuBr crystal by resonant Brillouin scattering.[17]

At higher temperature ($T\geq 40$ K), a strong deviation from the above fit is observed for the $\omega^b_{e\pi}$ polariton (the investigated temperature range is not sufficient to observe this deviation for the $\omega^f_{e\pi}$ polariton). This can be ascribed to LO-phonon-assisted mechanisms mediated by the Fröhlich interaction.[15] Here, only the up-conversion process needs to be taken into account because of the very low density of accessible states for the down-conversion mechanism (Fig. 2), and, hence,

$$\Gamma_{LO}(T)=\gamma_{LO}n(\omega_{LO}), \tag{6}$$

where $\omega_{LO}=26$ meV for CuCl and γ_{LO} is a frequency-dependent coupling constant. Including this mechanism,

$$\Gamma_{e\pi}=\Gamma_{LO}+\Gamma^{LA}_{DP}+\Gamma^{TA}_{PE}, \tag{7}$$

we obtain a temperature dependence in good agreement

with our experimental results using $\gamma_{LO}=26$ meV for the $\omega^b_{e\pi}$ polariton and a related value for the $\omega^f_{e\pi}$ polariton (solid lines in Fig. 3).

All the above coupling constants can be independently estimated using the known parameters in CuCl.[6] For the $\omega^f_{e\pi}$ polariton, the estimated DP and PE coupling factors are, respectively, ≈ 9 and ≈ 10 μeV. Their order of magnitude is in reasonable agreement with the values used to fit our experimental results (37 and 25 μeV, respectively). As a consequence of relations (4), a similar agreement is obtained for the $\omega^b_{e\pi}$ polariton. For this polariton, the experimental Fröhlich coupling constant γ_{LO} (=26 meV) is also comparable to the theoretical value (≈ 14 meV). This additional comparison between theory and experiment supports the above interpretation of the measurements pointing out the role of the acoustical and optical phonons in the polariton damping. Similar measurements were performed on the longitudinal mode in a (111) crystal. The results are comparable to those obtained for the $\omega^b_{e\pi}$ polariton and will be published in a forthcoming paper.

In conclusion, we have demonstrated that the dephasing of upper-branch exciton polaritons can be directly and selectively investigated by use of a time-resolved two-photon nonlinear technique. The results in CuCl reveal that the dephasing rate is dominated by population relaxation while pure dephasing through intraband mechanisms is negligible. The exciton-polariton relaxation is dominated by acoustical phonons at low temperature ($T \leq 40$ K) while the Fröhlich interaction becomes dominant for higher temperatures, corresponding to very different redistribution channels of the initial energy in the crystal.

Such experiments are readily applicable to other semiconductors allowing us to gain a precise map of the different exciton-polariton decay channels in the low-polariton-density regime. Measurements could also be performed at higher densities where polariton-polariton scattering becomes dominant, using, for example, a linear excitation followed by a nonlinear two-photon coherent detection.

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